



Case CO/2-22750

IN THE UNITED STATES PATENT OFFICE

IN RE APPLICATION OF:

ADALBERT BRAIG ET AL.

SERIAL NO.: 10/528136

FILED: SEPTEMBER 23, 2003

FOR: SYNERGISTIC UV ABSORBER COMBINATION

Art Unit: 1624

Examiner: Venkataraman Balasubramanian

COMMISSIONER FOR PATENTS

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DECLARATION UNDER 37 CFR 1.132

I, Markus Grob, a citizen of Switzerland, residing in Reinach, Switzerland, declare:

that I was awarded the degree of Diplomchemiker (in English: Chemist) by the Federal Institute of Technology of Zuerich, Switzerland, in 1989 and that I was awarded the degree of Doctor scientiae naturalium, by the Federal Institute of Technology of Zuerich, Switzerland, in 1993;

that I have been engaged in the Plastic Additive Department in the field of Additivation and Stabilization of plastics, mainly the stabilization for engineering resins for CIBA-GEIGY AG, Basel, Switzerland, now Ciba Specialty Chemicals Inc., Basel, Switzerland since 1995;

that I am familiar with the subject-matter of U.S. Patent Application No. 10/430128.

that I am familiar with the subject matter of the prior art US-B-6255483.

that the experiments described below have been made under my direction and supervision; and

that I am submitting herewith the following exact report of the tests made and the results obtained.

It was the object of the tests reported below to compare the yellowing after weathering of polymer compositions containing combinations of UV absorbers according to the present invention with polymer compositions containing the single UV absorbers of the state of the art.

### **Tested compounds**

component (A):

compound A2, page 9 of the description,

component (B):

compound v, page 5 of the description, a benzotriazole of formula (IIa); this compound is disclosed in US-B-6255483, col. 19, l. 5-6 and as formula 126 in col. 48 (example 26 of US-B-6255483)

compound xix, page 5 of the description, a 2-hydroxyphenyltriazine of formula (IIId); this compound is disclosed in US-B-6255483, col. 20, l. 58-59

compound xliii, page 7 of the description, a cinnamate of formula (IIe); this compound is encompassed by acrylate of US-B-6255483, col. 19, l. 28

Irgafos® 168: tris(2,4-di-tert-butylphenyl)phosphite

### **Formulation**

3.0 kg of a polycarbonate powder (dried for 8 hours at 120 °C in a vacuum drying oven (Lexan 145 resin 111 of General Electric)), Irgafos® 168 (0.08%) and the additives listed in table 1 are mixed for 2 minutes in a Henschel 5 l mixer. This mixture is then extruded in a Berstorff ZE 25x32D at a maximum temperature of 280 °C. The polymer strand is then granulated.

Using an injection molding machine, plates having a layer thickness of 2 mm are then molded from the granulate at a maximum temperature of 300 °C.

### **Weathering**

The samples are artificially weathered in accordance to ASTM G 53-96 on a QUV Tester (Q-Panel Company) equipped with 4 UV-B fluorescent lamps of UV-B type with a total irradiation of 0.63 W/m<sup>2</sup> at 313 nm with a cycle of 70 °C light period, 50 °C dark period BPT 8 hours dry and light followed by 4 hours water condensation and dark.

The yellowness index ( $YI_{\text{measured}}$ ) of these plates is then determined according to DIN 6167 and the DE value is measured in accordance to DIN 6174 after 1532 h of QUV-B irradiation. DE value is obtained by subtracting  $YI_{\text{measured}}$  prior to weathering from  $YI_{\text{measured}}$  after weathering.

### **Result**

A plate without UV absorber, plates with the compound A2, v, xix or xliii (state of the art) and plates with a combination of compounds A2 and v, a combination of compounds A2 and xix and a combination of compounds A2 and xliii (according to the invention) are compared in Table 1.

Table 1

UV absorber	YI <sub>measured</sub> <sup>*</sup>	YI <sub>calculated</sub>	DE <sub>measured</sub> <sup>*</sup>	DE <sub>calculated</sub>
-	46.4	-	28.8	-
0.30% A2 <sup>a</sup>	30.1	-	14.5	-
0.30% v <sup>a</sup>	32.6	-	18.2	-
0.30% xix <sup>a</sup>	29.4	-	15.3	-
0.30% xliii <sup>a</sup>	31.9	-	18.3	-
0.15% A2 <sup>b</sup> 0.15% v	27.7	31.4	14.1	14.9
0.15% A2 <sup>b</sup> 0.15% xix	27.2	29.3	13.8	14.2
0.15% A2 <sup>b</sup> 0.15% xliii	27.8	31.0	14.6	16.4

<sup>a</sup> State of the art

<sup>b</sup> according to the invention

\* Low values are advantageous

YI<sub>calculated</sub> and DE<sub>calculated</sub> indicate which values of YI<sub>measured</sub> and DE<sub>measured</sub> a skilled artisan expects if a combination of A2 and v, xix or xliii is used for the stabilisation of polymer compositions.

YI<sub>calculated</sub> and DE<sub>calculated</sub> are obtained according to the following formula:  
value (combination of 0.15% A2 and 0.15% B (v, xix or xliii) = 0.5\* value (0.30% A2) + 0.5\* value (0.30% B (v, xix or xliii))

There is a synergistic effect in case YI<sub>calculated</sub> and DE<sub>calculated</sub> are higher than YI<sub>measured</sub> and DE<sub>measured</sub>.

## **Conclusion**

These comparative experiments show that the combinations according to the invention exhibit a considerable synergistic effect compared to the single UV absorbers.

These comparative experiments provide evidence that YI<sub>measured</sub> of compositions according to the invention are considerably lower than YI<sub>measured</sub> of the state of the art compositions with a single UV absorber.

Moreover, these comparative experiments show that DE<sub>measured</sub> of compositions according to the invention are comparable to or lower than DE<sub>measured</sub> of the state of the art compositions with a single UV absorber.

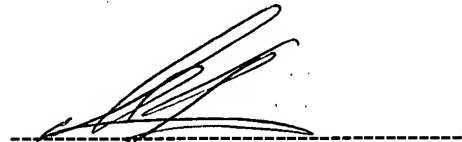
These findings are surprising and would not have been expected at the time this invention was made.

Hence, these comparative experiments provide evidence that the subject matter claimed in the present application is non-obvious in the light of the prior art.

**Final Statement**

I, Markus Grob, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Signed, this 8th day of August 2007

A handwritten signature in black ink, appearing to be 'M. Grob', is written over a horizontal dashed line.

Dr. Markus Grob